Inorganic Chemistry

Forum Article

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Preface: Forum on Aspects of Inorganic Chemistry Related to Nuclear Energy

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According to the U.S. Energy Information Administration's 2012 International Energy Outlook report,[1] global energy demands will grow by more than one-third through 2035. Because of the terawatt-scale increases in energy demand that are going to be required, it is imperative for humanity that we develop safe, sustainable, and reliable methods for global-scale energy production. In this regard, reports have appeared about the various roles that different sources of energy may play going forward (a so-called energy mix).[2] This special Forum issue of *Inorganic Chemistry* is dedicated to the particular subject of nuclear energy and highlights a number of areas in which chemistry, in particular inorganic chemistry, can improve technical issues surrounding nuclear power production.

From a historical perspective, the possibility of harnessing nuclear energy for electrical power generation emerged as a result of the discovery of neutron-induced fission in the late 1930s.[3] However, while the initial promise of electricity production via nuclear power was hinted at earlier in the 20th century, it was really not until after the second world war, and in particular during the late 1940s and early 1950s, that the use of nuclear energy for peaceful purposes rather than military applications came to the fore.[4]

There have been many contentious debates as to the pros and cons of nuclear power production, and some of these specific issues are highlighted in more detail within the contributions here. Proponents of nuclear energy hold the belief that it represents a sustainable, reliable, and economic method of large-scale electrical power generation that also contributes toward decreased CO_2 emissions. A recent publication reviews and analyzes nuclear power in this regard.[5] Other groups challenge the practice of nuclear power generation as something that presents a number of potential waste and environmental contamination concerns. There are certainly notable examples that illustrate this concern and the potential for release of radionuclides into the environment, including the 1979 Three Mile Island accident and the disasters in Chernobyl (1986) and Fukushima Daiichi (2011). Concerns about the possibility of nuclear weapons proliferation have also arisen as a result of nuclear energy production.[6]

As some background for the contributions that follow in this particular issue, a brief overview of some of the processes related to nuclear power production is warranted. Within a nuclear reactor, a controlled (or moderated) self-sustaining nuclear chain reaction is initiated and used to generate thermal energy. Several classes of nuclear reactors are currently used for large-scale electrical power generation, and these are characterized by the type of fuel, coolant, moderator, etc., that they utilize.[7] The heat generated by fission within the reactor core can be passed to some sort of working fluid (for example, water or a gas), which then drives turbines that can be used to turn electrical generators or to drive other mechanical components such as propellers for marine propulsion applications.

The efficacy of nuclear power production rests on the ability to effect a so-called nuclear fuel cycle safely, efficiently, and economically. "Fuel cycle" is the terminology used to describe how the nuclear fuel itself progresses through the various different stages involved in generating electricity. A particular type of fuel cycle thus comprises steps in the so-called "front end", which represent the preparation of the fuel (e.g., mining and fuel enrichment), the "service period" during which time the fuel material is used in reactor operation for electricity production, and the "back-end" steps, which are aimed toward the safe handling and subsequent reprocessing or disposition of spent nuclear fuel materials. If used fuel remains unprocessed and is simply sent to a repository; the fuel cycle is called an "open fuel cycle" (sometimes referred to as a "once-through cycle").[8] Correspondingly, if the used fuel is reprocessed, it is referred to as a "closed fuel cycle". While some countries have adopted the use of nuclear power to produce a significant percentage of their electricity (e.g., the U.S., France, Japan, and others), many have chosen not to pursue this as an option (e.g., Australia and several European countries). Some of the nations that have invested in a nuclear power infrastructure have also decided to reprocess spent fuels (including some countries in Europe and Japan), while others do not practice reprocessing (e.g., the U.S.) because of the desire to prevent nuclear proliferation. Some specific technical challenges and requirements associated with efficacious "back-end" solutions are alluded to in more detail later in the Forum.

For the sake of consistency, contributions within the Forum have been grouped into several common themes that relate to different aspects of the fuel cycle. The first set of papers focuses on the general area of separations and extraction chemistries. These are of relevance to various schemes aimed toward the reprocessing of spent fuels. One of the emerging themes in this particular area is that covalency (a notion central to the discipline of chemistry) can potentially be exploited to effect the separation of components contained within used fuel materials. In the first contribution from the U.K., Nik Kaltsoyannis highlights various computational approaches that have been used to probe the relative degree of covalency within

Special Issue: Inorganic Chemistry Related to Nuclear Energy

Received: March 12, 2013
Published: April 1, 2013

dx.doi.org/10.1021/ic4006136 | *Inorg. Chem.* 2013, 52, 3405−3406

lanthanide and actinide complexes. The following two articles from Michael Hudson and co-workers and Clint Sharrad and co-workers highlight coordination chemistry pertinent to spent-fuel materials separation technologies, in each case describing the use of nitrogen-donor-ligand systems. The paper from Anne Gorden and colleagues here in the U.S. reviews some recent efforts aimed at understanding some of the fundamental chemistry differences between the 4f and 5f elements, again following the general theme of allowing better extraction schemes to be developed. Yasushisa Ikeda and colleagues in Tokyo then describe some chemistry of the actinide elements in ionic liquids (ILs). The focus here is on complex formation, the electrochemical behavior of uranyl species in ILs, and extraction studies of uranyl complexes. The last contribution describes the application of chemistry to the area of nuclear waste separations. Bruce Moyer and colleagues in the U.S. describe the application of molecular recognition strategies for the removal of sulfate from radioactive wastes and describe some of the benefits of doing so.

A paper from Europe from Vincenzo Rondinella and co-workers then focuses on some of the technical challenges related to spent-fuel materials that will be buried in a geologic repository. In particular, studies aimed toward an understanding of issues surrounding corrosive behavior are described. A series of papers related to the environmental chemistry of the actinides are then highlighted. The first of these by Kate Maher and colleagues at Stanford and SLAC National Accelerator Laboratory in the U.S. reviews the speciation chemistry of the actinide elements and alludes to a variety of spectroscopic techniques used to quantify and predict microscopic to macroscopic transport behaviors of these elements. Annie Kersting from Lawrence Livermore National Laboratory (LLNL) then describes some aspects of plutonium environmental chemistry with some emphasis on the role that colloidal forms of this element play in its mobility in the environment. A paper from Dave Clark and colleagues at Los Alamos National Laboratory (LANL) completes this section. Herein the use of various spectroscopic techniques are used to probe the nature of neptunium chemistry under highly alkaline conditions pertinent to alkaline radioactive waste forms.

The final three contributions focus on fundamental molecular chemistry of elements pertinent to the nuclear fuel cycle. The first of these from Justin Walensky, Trevor Hayton, and co-workers describes some new efforts in actinide alkyl chemistry, and that is followed by a manuscript from Bill Evans and colleagues describing metal cyclopentadienyl chemistry. The last manuscript from Al Sattelberger and Ken Czerwinski addresses some chemistry of the lightest radioelement, technetium. These authors allude to how an understanding of the chemistry of this element and the ability to manipulate its chemistry can be exploited in the fuel cycle for waste forms and in separation technologies.

In testimony to the magnitude of the challenges facing us in energy production, storage, and utilization, Dr. Peter Lyons, the Assistant Secretary for Nuclear Energy at the Department of Energy stated, "The Energy Department is committed to an all-of-the-above energy strategy that develops every source of American energy, including nuclear power. Our efforts to restart the nuclear energy industry include advancing nuclear chemistry and technologies that will make nuclear power safer and more efficient. That commitment is also demonstrated by our Nuclear Energy University Program, which funds research and development for cutting-edge nuclear energy technologies at American universities and colleges, helping to train and educate the next generation of leaders in the U.S. nuclear industry."[9]

It is indeed apparent that energy is a tremendous challenge facing humanity in the 21st century, and exactly what role nuclear energy will play in that future picture remains to be seen. Unquestionably inorganic chemistry, and inorganic chemists, will play an important role in shaping energy research and developments and in providing solutions to these grand challenges.

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Notes

The authors declare no competing financial interest.

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(7) See http://www.world-nuclear.org/info/Nuclear-Fuel-Cycle/Power-Reactors/Nuclear-Power-Reactors (the World Nuclear Association, 2012).

(8) For stages of the nuclear fuel cycle, see http://www.nrc.gov/materials/fuel-cycle-fac/stages-fuel-cycle.html.

(9) Lyons, P. , Assistant Secretary for Nuclear Energy at the Department of Energy. Personal communication from Karissa Marcum, Office of Public Affairs to Dr. Tamara Hanna, Managing Editor, *Inorganic Chemistry*.